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# Ultra-high critical current densities, the vortex phase diagram and the effect of granularity of the stoichiometric high- $T_c$ superconductor, $\text{CaKFe}_4\text{As}_4$

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We present a comprehensive study of the critical current densities and the superconducting vortex phase diagram in the stoichiometric superconductor  $\text{CaKFe}_4\text{As}_4$  which has a critical temperature of  $\sim 35$  K. We performed detailed magnetization measurements both of high quality single crystals for different orientations in an applied magnetic field up to 16 T and for a powder sample. We find an extremely large critical current density,  $J_c$ , up to  $10^8$  A/cm<sup>2</sup> for single crystals when  $H\parallel(ab)$  at 5 K, which remains robust in fields up to 16 T, being the largest of any other iron-based superconductor. The critical current density is reduced by a factor 10 in single crystals when  $H\parallel c$  at 5 K and significantly suppressed by the **presence** of grain boundaries in the powder sample. We also observe the presence of the fishtail effect in the magnetic hysteresis loops of single crystals when  $H\parallel c$ . The flux pinning force density and the pinning parameters suggest that the large critical current could be linked to the existence of point core and surface pinning. Based on the vortex phase diagram and the large critical current densities,  $\text{CaKFe}_4\text{As}_4$  is now established as a potential iron-based superconductor candidate for practical applications.

## I. INTRODUCTION

The discovery of iron based superconductors<sup>1,2</sup> has led to important advances in superconductivity not only because of their unconventional superconducting mechanism with rather high superconducting transition temperatures, but also due to their potential for high magnetic field applications, such as superconducting wires and tapes. The bulk iron-based superconductors have a high transition temperature of up to 58 K<sup>3,4</sup> and very high upper critical field ( $H_{c2}$ ), up to 100 T<sup>5,6</sup>. Furthermore, their high upper critical field, small anisotropy, good grain boundary connectivity and high critical current density under high magnetic field are all important factors contributing to their suitability for practical applications<sup>7,8</sup>.

Among iron-based superconductors, the recently discovered  $\text{CaKFe}_4\text{As}_4$  is a member of a new 1144 structural family of iron-based superconductors, being a stoichiometric optimally-doped superconductor, with a transition temperature,  $T_c$ , around 35 K and large upper critical field up to 90 T<sup>9,10</sup>. A number of transport, thermal and thermodynamics experiments on  $\text{CaKFe}_4\text{As}_4$  do not show any structural transition or magnetic order<sup>9,10</sup>. **This system** displays similar superconducting properties to those of the optimally-doped  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  (BaK122) system<sup>11</sup>.  $\text{CaKFe}_4\text{As}_4$  has a tetragonal structure ( $P4/mmm$ ), where Ca and K layers stack alternatively across the  $\text{Fe}_2\text{As}_2$  layer along the  $c$  axis<sup>9,10</sup>; Fe-As bond distances are different for the As atoms above and below the Fe plane, in contrast to the 122 system that has equivalent Fe-As distances. The lowering in symmetry has significant consequences on the electronic structure of  $\text{CaKFe}_4\text{As}_4$ , being that of a compensated metal with a large number of electronic bands close to a nesting instability<sup>12</sup> as well as on its magnetic structure which forms an unusual spin-vortex crystal order by Ni or Co doping<sup>13</sup>.

Generally, iron-based superconductors possess a layered

structure similar to that of the high- $T_c$  cuprates. Superconductivity **in these systems** often appears out of an antiferromagnetic state with bad metallic behaviour, found in the parent compounds, which is suppressed by electron (or hole) doping<sup>2,14,15</sup> or by application of external pressure<sup>2</sup>, giving rise to complex superconducting phase diagrams<sup>2,14,15</sup>. The understanding of the superconducting mechanism of unconventional superconductors is challenging due to different competing electronic phases, but the existence of few stoichiometric iron-based superconductors provides a unique opportunity to study this phenomenon in the absence of additional extrinsic effects introduced by chemical substitutions.

The critical current density,  $J_c$ , is an essential property of the high- $T_c$  superconductors in order to identify the best candidates for practical applications. A high  $J_c$  exceeding  $10^5$  A/cm<sup>2</sup> even under high magnetic field above 10 T is found in many optimally-doped iron-based superconductors<sup>7</sup>. The high transition temperatures, large magnetic penetration depth, vortex motion and thermal fluctuation generally plays an important role in the critical current properties, similar to the high- $T_c$  cuprates. These phenomena cause interesting effects in the vortex dynamics, such as giant-flux creep, thermally activated flux flow and **the presence of a second magnetization peak or the fish-tail effect**<sup>16,17</sup>. The ability to carry the critical current is governed by the vortex pinning strength and motion. Different kinds of pinning determine the elastic or plastic motion of vortex lattice, formation of vortex glass, vortex melting and order-disorder phase transitions<sup>17,18</sup>. As vortex motion is strongly affected by the effects of disorders in superconductors, it is important to understand what happens in a stoichiometric and clean high- $T_c$  superconductor, such as  $\text{CaKFe}_4\text{As}_4$ .

In the present study, we report the superconducting properties and the critical current densities of  $\text{CaKFe}_4\text{As}_4$  in both single crystals and polycrystalline samples using magnetization measurements in magnetic fields up to 16 T. We find that

CaKFe<sub>4</sub>As<sub>4</sub> exhibits an ultra-high critical current density of  $J_c \sim 10^8$  A/cm<sup>2</sup> at 5 K, when the magnetic field is parallel with the conducting plane, as extracted from isothermal magnetic hysteresis loops. Critical current density is reduced by a factor 10 in single crystals, **when the magnetic field is parallel with the *c*-axis at 5 K**, and it is significantly suppressed by the effect of grain boundaries in powder samples. Our study compares the behaviour of the lower critical field ( $H_{c1}$ ) and the upper critical field ( $H_{c2}$ ) of single crystals and the vortex phase diagram of single and polycrystalline samples. We detect the presence of a second magnetization peak in the magnetic hysteresis loops of single crystals when  $H \parallel c$ . The flux pinning force density and the pinning parameters suggest the existence of surface and point core pinning of vortices. Our results suggest that CaKFe<sub>4</sub>As<sub>4</sub>, with such large critical current densities and very high upper critical fields, is an exciting new candidate for practical applications.

## II. EXPERIMENTAL DETAILS

The single crystal CaKFe<sub>4</sub>As<sub>4</sub> samples were grown by the FeAs self-flux method and polycrystalline sample were synthesized by solid state reaction method, as detailed elsewhere<sup>9,10,19</sup>. The single crystals have rectangular shapes for both magnetic and resistivity measurements, whereas the polycrystalline sample was pressed powder in a cylindrical shape. Magnetic measurements were carried out using the VSM (Vibrating Sample Magnetometer) option of a Physical Property Measurement System (PPMS-Quantum Design) with the magnetic fields up to 16 T. The magnetic isotherms were recorded up to 16 T with ramping rate of 3 mT/s, at several temperatures ranging from 2 up to 40 K. All the hysteresis loops at constant temperatures,  $M(H)$ , and temperature dependent magnetization in constant magnetic field,  $M(T)$ , were performed after cooling the sample in zero magnetic field from above  $T_c$ .

Magnetization hysteresis loops allow us to extract the critical current density,  $J_c$ , as a function of magnetic field using the extended Bean's critical state model<sup>20</sup>. The vortex pinning force density can be estimated using  $F_P = J_c \times \mu_0 H$ <sup>21</sup>. For a rectangular crystal, the magnetic field dependence of the critical current density,  $J_c$  (in units of A/cm<sup>2</sup>) is given by the relation  $J_c = 2\Delta M/[a(1-a/3b)]$  (for  $a < b$ );  $\Delta M = M_{up} - M_{dn}$ , and  $M_{up}$  and  $M_{dn}$  are the magnetization while magnetic fields are decreasing and increasing, respectively. The magnetization  $\Delta M$  expressed in A/cm is obtained from the magnetic moment divided by the sample volume,  $V = a \times b \times c$ , where  $a$  and  $b$  are in-plane sample dimensions perpendicular to the applied magnetic field and  $c$  is sample dimension, parallel to the applied magnetic field. We have measured a single crystal of CaKFe<sub>4</sub>As<sub>4</sub> with the following dimensions:  $a = 1.09$  mm,  $b = 1.365$  mm and  $c = 0.0025$  mm, measured with the magnetic field aligned along  $c$ -axis ( $H \parallel c$ ) and  $a = 0.0025$  mm,  $b = 1.09$  mm and  $c = 1.365$  mm, measured with the magnetic field aligned in the  $ab$ -plane ( $H \parallel (ab)$ ). The sample thickness of the crystal was evaluated using Gaussian fits to the different optical image measurements across the sample, as shown in

Fig.SM1. For the cylindrical pressed powder sample in order to estimate the critical current,  $J_c$ , we used the expression  $J_c = 3 \Delta M/d$ , where  $d = 5.03$  mm and  $l = 4.08$  mm are the diameter and length of the cylinder, respectively<sup>20</sup>. **The average grain size for the powder sample is around 1.7(7)  $\mu$ m.**

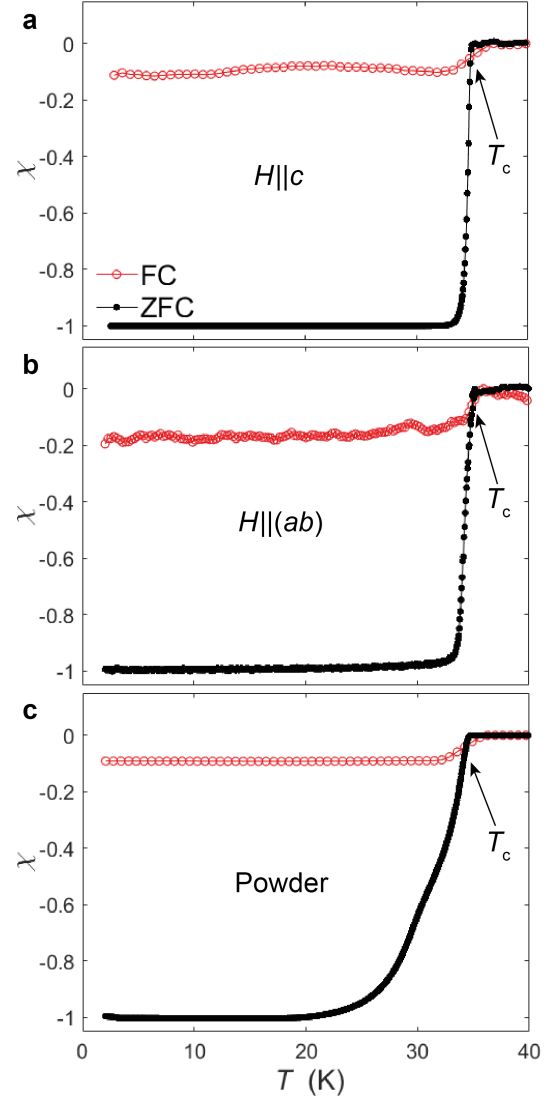


FIG. 1. Temperature dependence of magnetic susceptibility ( $\chi$ ) deduced from zero-field cooled and field-cooled magnetization measurements for single crystal when (a)  $H \parallel c$  and (b)  $H \parallel (ab)$  in 5 mT as well as (c) the powder sample in 2 mT. The arrows indicate the positions of the onset values of the critical temperature,  $T_c$ .

To estimate the lower critical field,  $H_{c1}$ ,  $M(H)$  loops were measured up to 0.5 T sweeping the field with a slow rate of 0.2 mT/s at each temperature. Before each measurement the sample was warmed up above  $T_c$ , and the remnant field of the magnet is reduced before cooling down to the target temperature. The susceptibility data,  $\chi = M/H$  has been corrected for the demagnetization effect<sup>22</sup>. For an applied external field  $H_a$ , the internal field  $H_i = H_a - N_{\text{eff}}M$ , where  $N_{\text{eff}}$  is the effective demagnetization factor. For the accurate determination of  $H_{c1}$ , we subtracted the value of magnetization obtained by

the low-field magnetization slope from the magnetization ( $M$ ) for each isotherm<sup>23</sup>. The deviation point of magnetization versus field from linear behaviour gives the value of the first penetration field,  $H_{c1}^*$ . We estimate the effective demagnetization factor for our single crystal to be  $N_{\text{eff}} = 0.995$  when  $H||c$ , which is similar to the calculated demagnetization factor for this orientation<sup>22</sup>. When  $H||(ab)$  the value of  $N_{\text{eff}} = 0.70$  which is extracted from zero-field cooled data assuming that in the orientation the sample is a perfect diamagnet as for  $H||c$ .  $N_{\text{eff}} = 0.41$  for the cylindrical polycrystalline sample is extracted both from calculations and experiment<sup>22,24</sup>. The upper critical field,  $H_{c2}$ , is extracted both from the temperature dependence of magnetization at different constant magnetic field up to 16 T.

### III. RESULTS AND DISCUSSION

The temperature dependence of the magnetic susceptibility ( $\chi$ ) from zero-field-cooled (ZFC) and field-cooled (FC) magnetization measurements in low magnetic field (2 or 5 mT) for single crystal (when  $H||c$  and  $H||(ab)$ ) and for the powder sample of  $\text{CaKFe}_4\text{As}_4$  are shown in Fig. 1(a)-(c). A perfect diamagnetic state is reached at low temperatures for all measured samples. The critical temperature,  $T_c$ , is defined around 35 K from the bifurcation point between ZFC and FC branches of the magnetization in Fig. 1. The width of the superconducting transition is very narrow for the single crystal ( $\Delta T_c \sim 1$  K) suggesting a high quality of this sample, as shown by the X-ray and resistivity data in the Supplemental Material in Fig. SM1. The powder sample also show perfect diamagnetism at low temperatures but it has a much broader transition of ( $\Delta T_c \sim 12$  K), likely due to pinning to grain boundaries and grain size effects, as seen in other iron-based superconductors<sup>25</sup>. No secondary phases were identified in our powder sample, as shown in Fig. SM2. The small diamagnetic signal in the FC data reflects the strong pinning nature of the sample and permanent flux trapped inside it.

The temperature dependence of the superconducting lower critical field,  $H_{c1}$ , is determined using low-field magnetic hysteresis measurements, as shown in Fig. 2(a)-(c). We measured the field dependence of the magnetization in the superconducting state at different temperatures, with the external field in the  $(ab)$ -plane and along the  $c$ -axis and also for the powder sample. The linear variation of the magnetization is a signature of the Meissner state up to  $H_{c1}$ , above which the vortices starts to form. The temperature dependence of the extracted  $H_{c1}$  for the single crystal and the powder sample is shown in Fig. 2(d). The values of  $H_{c1}$  for  $H||(ab)$  is higher than that for  $H||c$ . The  $H_{c1}$  value for polycrystalline sample lies in between these two orientation of the single crystal (as shown in Fig. 2d), as expected from the powder averaging effects, and it was extracted by fitting a linear dependence to the magnetization above the anomaly caused by the grain boundaries. **The  $H_{c1}(T)$  powder data show a very weak temperature dependence at low temperatures below 10 K, consistent with recent  $\mu\text{SR}$  results<sup>26</sup>.**

To determine the upper critical field,  $H_{c2}$ , we have mea-

sured zero-field cooled (ZFC) magnetization in the temperature range between 2 to 40 K for various values of the magnetic field up to 16 T, shown in Figs. 2(f)-(h) for the single crystal and the powder sample, respectively. The upper critical field,  $H_{c2}(T)$ , as a function of temperature near  $T_c$  is defined by the value at which magnetization starts to deviate from the high-temperature paramagnetic value. Just below the critical temperature, we noticed a small jump in the temperature dependence of zero-field magnetization for both the single crystal and the powder sample, being more pronounced as the field increases. This effect may be caused by the flux jumps.

Fig. 2(i) shows the  $H - T$  phase diagram of  $\text{CaKFe}_4\text{As}_4$  calculated from magnetization data. Since the transition temperature does not shift much **with magnetic field towards low temperatures**, it indicates a very high value of  $H_{c2}(0)$ . To estimate  $H_{c2}(0)$  we use the Werthamer-Helfand-Hohenberg (WHH) formula<sup>27</sup> close to  $T_c$ ,  $\mu_0 H_{c2}(0) = -0.69T_c(d\mu_0 H_{c2}/dT)$ . The slope of upper critical field ( $-d\mu_0 H_{c2}/dT$ ) (see Table SM1) ranges between 2.6(1) for  $H||c$ , to 5.7(7) for  $H||(ab)$  for the single crystal and 10.1(5)  $\text{TK}^{-1}$  for the powder sample. Using the transition temperature as  $T_c = 35$  K, we find  $\mu_0 H_{c2}$  at zero temperature of  $\sim 62$  T when  $H||c$  and a much larger value of 135 T when  $H||(ab)$ . The powder sample show a significantly larger upper critical field of 241 T, a factor of  $\sim 4$  larger than in single crystals. We have also measured the resistivity for a different single crystal, as shown in Fig. 1SM(c). The slope of upper critical field from transport data measured in magnetic field (not shown here<sup>28</sup>) gives much larger upper critical fields,  $\mu_0 H_{c2}(0) = 82$  T, and 168 T for  $H||c$  and  $H||(ab)$ , respectively, similar to those reported previously<sup>10</sup>.

Using the zero temperature value of upper critical field  $H_{c2}(0)$ , the corresponding value of  $\mu_0 H_{c2}(0)/k_B T_c$  is close to 1.8 for  $H||c$ , and increases to 3.9 when  $H||(ab)$  and 6.9  $\text{TK}^{-1}$  for the powder sample. The Pauli limit is reached when the condensation energy is overcome by the Zeeman energy for normal electrons, defined as  $\mu_0 H_p/T_c = 1.84 \text{ T K}^{-1}$  in the case of singlet pairing and weak spin-orbit coupling<sup>29,30</sup>. In order to estimate the superconducting order parameter we used the Ginzburg-Landau (GL) approach. This gives a coherence length extrapolated to zero temperature as  $\xi_{ab}=1.9$  nm,  $\xi_c=0.87$  nm for the single crystal (as shown in Fig. 2j) and  $\xi = 0.97$  nm for the powder sample. The extracted mean free path is  $\ell = 15.7$  nm (using carrier concentration reported in Ref. 10) based on the extrapolated normal resistivity at zero temperature of  $\rho_n \sim 15.9 \mu\Omega\text{cm}$  (see Fig. SM1(c)). This suggests that our crystals lie within the clean limit with  $\xi(0) \ll \ell$ .

The  $H_{c1}$  values extracted in Fig. 2(d) were used to calculate the London penetration depth as a function of temperature,  $\lambda(T)$ . In order to estimate the penetration depth for the powder sample, we correct the value for demagnetizing effects (as detailed earlier using  $N_{\text{eff}}=0.41$ ) and estimate the value of the Ginzburg-Landau parameter as  $\kappa \sim 88$ . This gives a low temperature London penetration depth  $\lambda(0)$  of 104 nm (as shown in Fig. 2e), which is similar to the value of 133 nm, extracted from London penetration depth studies in single crystals<sup>31</sup>. Muon relaxation studies estimate the pen-

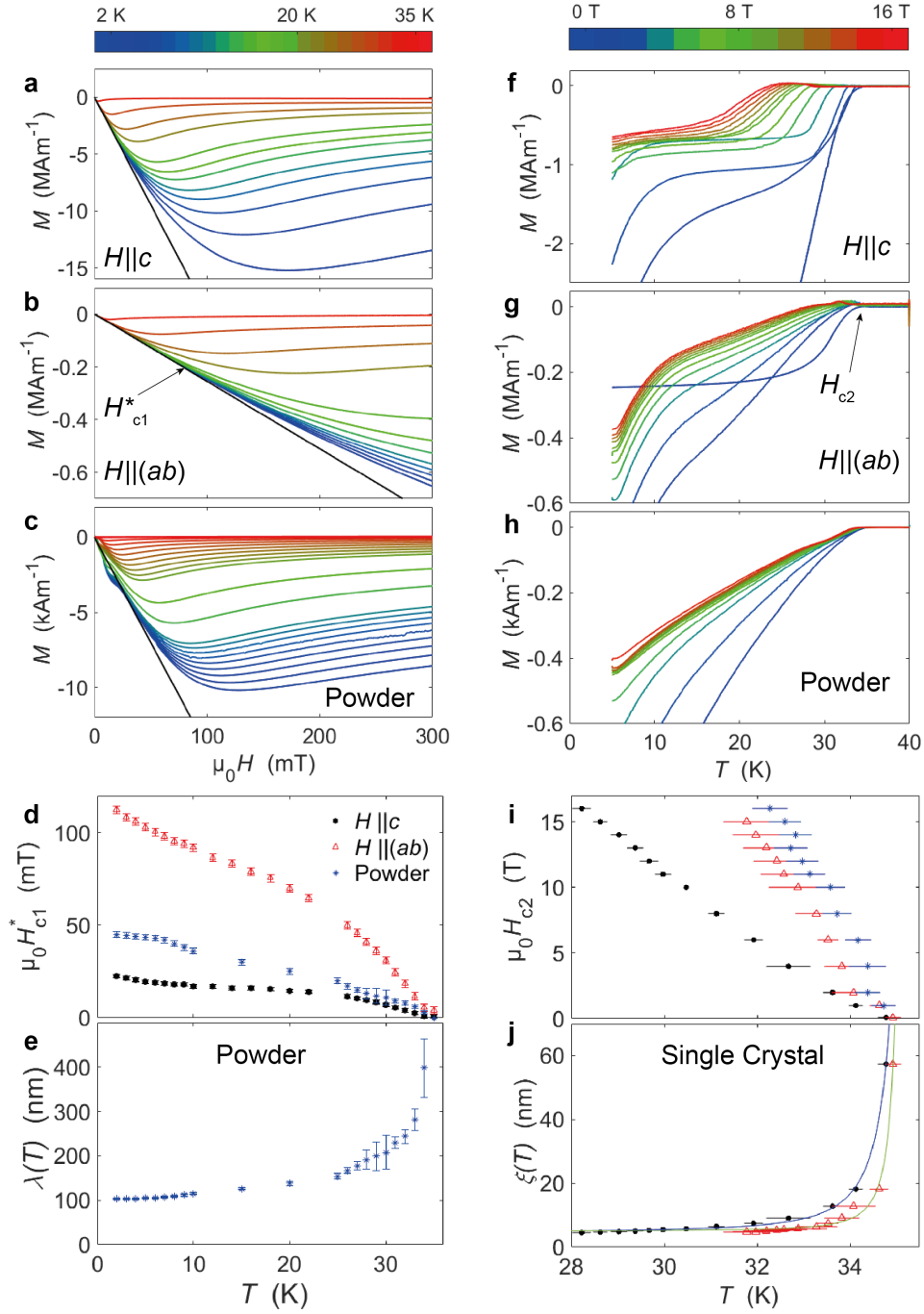


FIG. 2. Magnetization,  $M$ , versus magnetic field at constant temperatures for the single crystal when (a)  $H||c$  and (b)  $H||(ab)$  as well as for (c) the powder sample of  $\text{CaKFe}_4\text{As}_4$ . The arrow in (b) shows the point of deviation of magnetization from linear dependence in the Meissner state, defined as the first penetration field  $H_{c1}^*$ . (d) The temperature dependence of the lower critical field  $H_{c1}^*$  measured for the single crystal in two orientations in magnetic field as well as the powder sample. (e) The penetration depth,  $\lambda$ , for the powder sample. **The temperature dependence of magnetization in different magnetic fields up to 16 T for the single crystal in two orientations in (f)-(g) and for the powder sample in (h).** (i) The upper critical field  $H_{c2}$  versus temperature for the single crystal and the powder sample. (j) The coherence lengths,  $\xi_{ab}$  and  $\xi_c$ , for a single crystal of  $\text{CaKFe}_4\text{As}_4$ .

etration depth as 209 nm for a single crystal<sup>32</sup> and 286 nm for a powder sample<sup>26</sup>.

Figs. 3(a-c) show the  $M(H)$  loops at constant temperatures between 2 to 35 K in a magnetic field up to 16 T for

the single crystal in the two orientations ( $H||(ab)$  and  $H||c$ ) and for the powder sample, respectively. The hysteresis areas decreased with increasing temperature and the symmetrical shape of  $M(H)$  loops imply the existence of flux pin-

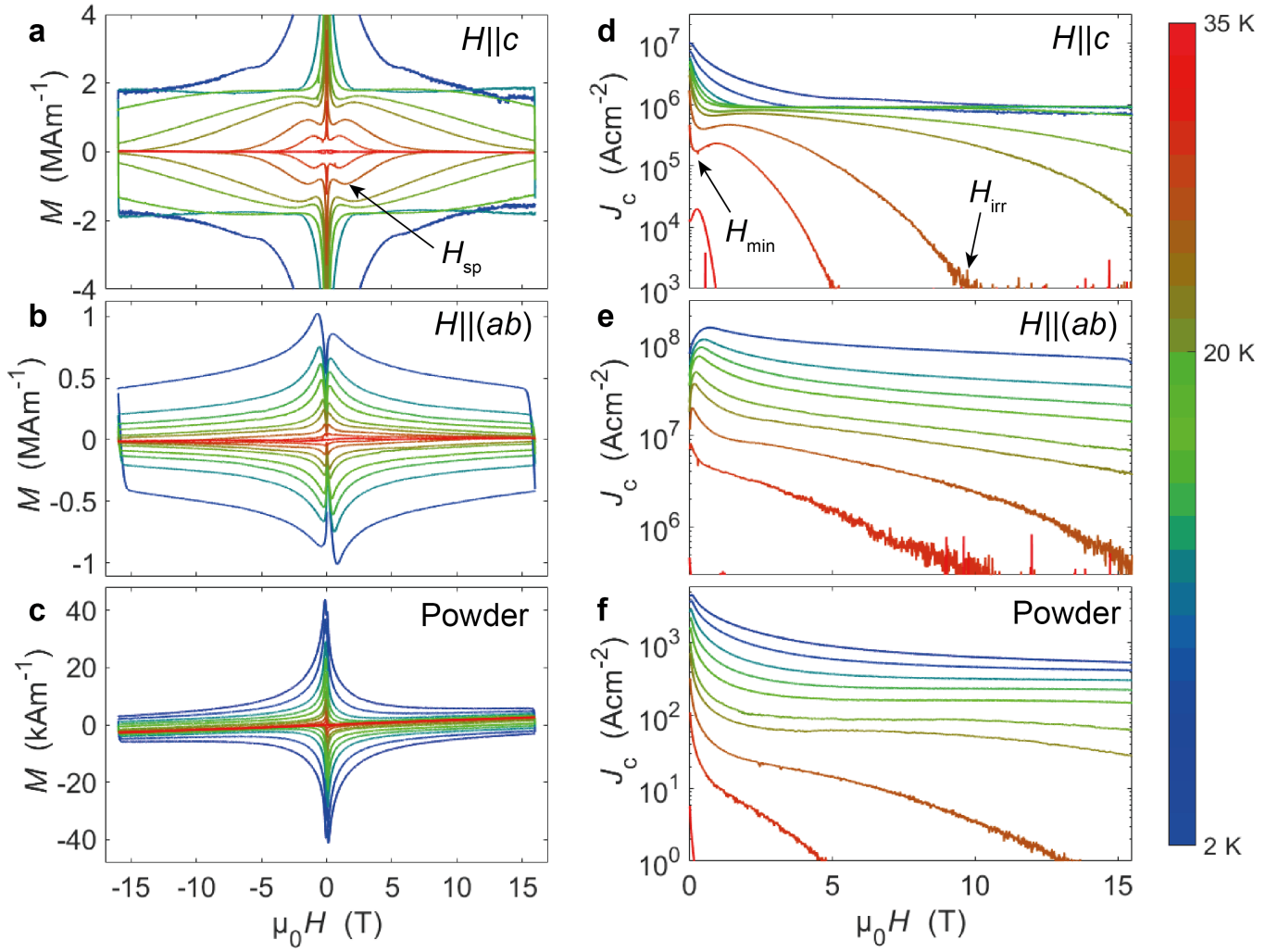


FIG. 3. The isothermal magnetization loops,  $M(H)$ , for the single crystal in two orientations with (a)  $H||c$  and (b)  $H||(ab)$  and (c) the powder sample of CaKFe<sub>4</sub>As<sub>4</sub>. The critical current density,  $J_c$ , calculated from  $M(H)$  using the Bean model<sup>20</sup>, plotted as a function of magnetic field at constant temperatures for the single crystal (d) when  $H||c$  and (e)  $H||(ab)$  and (f) the powder sample, respectively. The arrows indicate the positions of  $H_{sp}$  in (a) and  $H_{irr}$  and  $H_{min}$  in (d).

ning centres, suggesting that the magnetization is dominated by bulk pinning rather than surface and geometrical barriers<sup>21</sup>. Furthermore, the magnetic hysteresis loop in Figs. 3(b) when  $H||(ab)$  displays a dip near zero magnetic field which may be caused by the highly inhomogeneous field distribution in the vortex state<sup>41</sup> and the anisotropy of  $J_c$ <sup>42</sup>.

The second magnetization peak (SMP) can be easily seen for the single crystal below 26 K when  $H||c$ , as shown in Fig. 3(a). With decreasing temperature, the peak moves to higher magnetic fields beyond the maximum applied magnetic field of 16 T. In the region between the valley and the peak, the width of the irreversible magnetization,  $\Delta M$ , in Fig. 3(a) extends with increasing magnetic field and shows a clear fish-tail effect. This effect was observed in other superconducting single crystals, such as cuprates<sup>43,44</sup>, Nb<sub>3</sub>Sn<sup>45</sup>, MgB<sub>2</sub><sup>46</sup> and iron-based superconductors, such as BaK122<sup>34</sup> and LiFeAs<sup>36</sup>. Although its origin is not fully explained, the fish-tail effect is strongly dependent on the sample orientation in externally ap-

plied magnetic field, being diminished when  $H||(ab)$ . Importantly, the width of the  $M(H)$  loops for  $H||(ab)$  (Fig. 3(b)) is much larger than of the loops for  $H||c$  (Fig. 3(a)) or the powder sample (Fig. 3(c)), implying that the pinning mechanisms is anisotropic and strongly affected by the granularity. Similar effects caused by anisotropy of flux pinning were observed in other iron-based superconductors, such as Fe(Se,Te)<sup>35</sup> and SmFeAs(O,F)<sup>33</sup>.

According to the extended Bean model<sup>20</sup>, the critical current density ( $J_c$ ) can be determined from the hysteresis loop width ( $\Delta M$ ). As the critical current remains rather constant with the change of magnetic field for the low temperature magnetization isotherms, the extended Bean's model gives a good estimation of the critical current density in our case. Other models that take into account the field variation of the critical current density and the vortex dynamics were also developed by Kim in Ref. 47 and Kim-Anderson and in Ref. 48. Figs. 3(d-f) show the field dependence of  $J_c$  at constant tem-

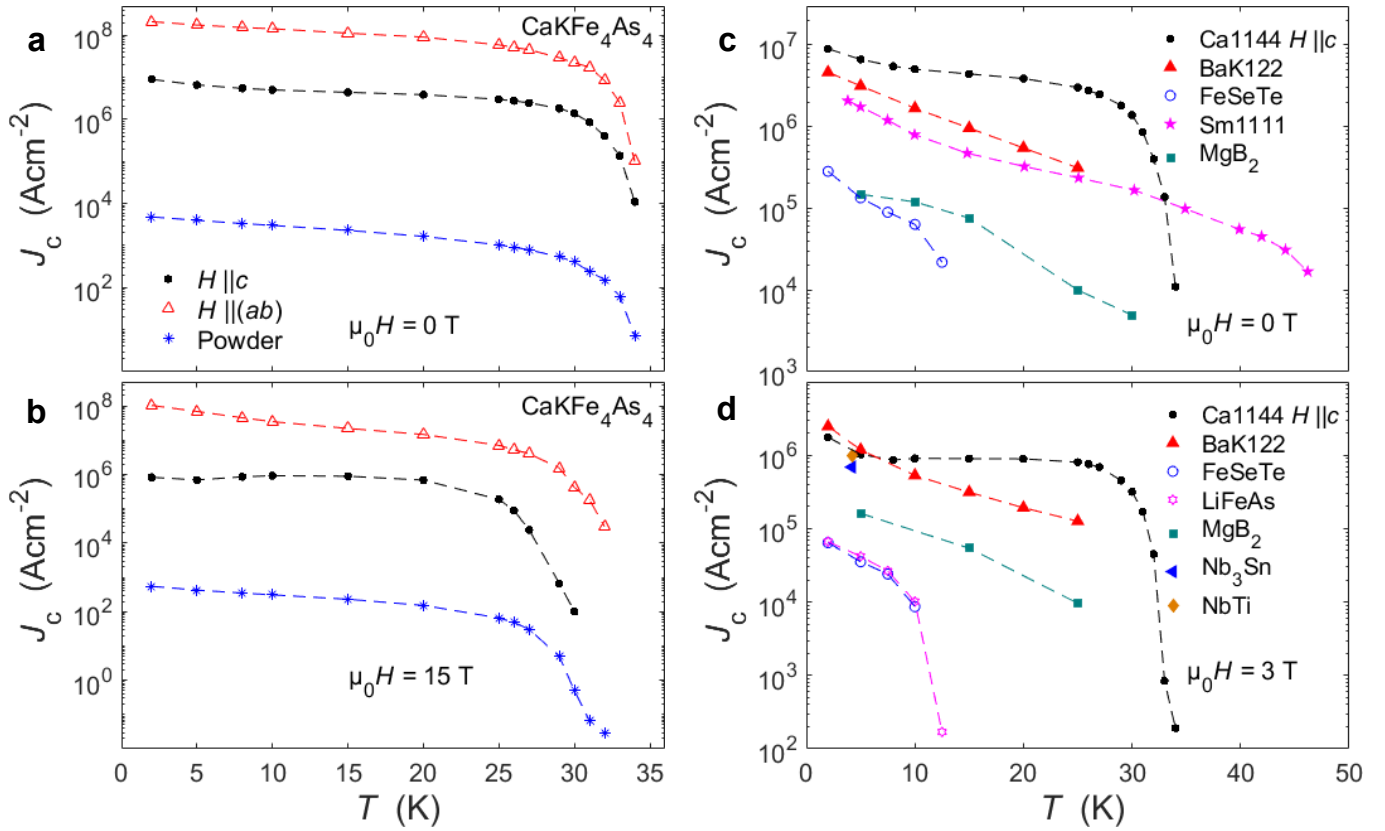


FIG. 4. The temperature dependence of critical current density,  $J_c$ , extrapolated at (a)  $\mu_0 H = 0$  T and (b)  $\mu_0 H = 15$  T of the single crystal in the two orientations in magnetic field and of the powder sample of  $\text{CaKFe}_4\text{As}_4$ . Comparison between the temperature dependence of the critical current density  $J_c$  (calculated using the Bean model<sup>20</sup>) in single crystals of  $\text{CaKFe}_4\text{As}_4$  with those reported for different iron-based superconductors ( $\text{SmFeAs(O,F)}$ <sup>33</sup>,  $(\text{Ba,K})\text{Fe}_2\text{As}_2$ <sup>34</sup>,  $\text{Fe(Se,Te)}$ <sup>35</sup>,  $\text{LiFeAs}$ <sup>36</sup>) and conventional superconductors ( $\text{MgB}_2$ <sup>37-39</sup>,  $\text{Nb}_3\text{Sn}$ <sup>39</sup>,  $\text{NbTi}$ <sup>40</sup>) in magnetic field of (c) 0 T and (d) 3 T for  $H \parallel c$ .

peratures obtained for the single crystal for different orientations in magnetic field and for the powder sample, respectively. The value of  $J_c$  for  $H \parallel (ab)$  has ultra-high values of the order of  $10^8$   $\text{A/cm}^2$ , which is an order of magnitude larger than the value obtained when  $H \parallel c$ . However, the powder sample has a  $J_c$  which is at least three orders of magnitude lower than that of the single crystal (Table SM1), but similar to other polycrystalline iron-based superconductors<sup>7,8</sup>. The low value of  $J_c$  of the powder sample is not surprising because the magnetic behaviour is significantly affected by the extrinsic factors, such as grain morphology, surface roughness, inter-grain voids and interfaces<sup>7</sup>. These extrinsic factors could be reduced by improving the synthesis process by adopting a ball-milling process, re-sintering and the optimization of both the temperature and length of the preparation process.

Fig.3(d)-(e) shows that  $J_c$  of  $\text{CaKFe}_4\text{As}_4$  is robust and little affected by a magnetic field up to 16 T, reaching a value of  $\sim 1.9(2) \times 10^8$   $\text{A/cm}^2$  for  $H \parallel (ab)$  for 1 T and only dropping to  $\sim 0.7(2) \times 10^8$   $\text{A/cm}^2$  by 15 T. These are some of the highest values obtained for any iron-based superconductor single crystal. This critical current value suggests strong vortex pinning, establishing this stoichiometric high- $T_c$  compound as a potential candidate for practical applications. To

further emphasise this important aspect, we have plotted  $J_c$  value for single crystal and the powder sample with respect to temperature for different magnetic fields of 0 and 15 T, as shown in Figs.4(a) and (b), respectively. We find that  $J_c$  remains large over a large temperature range between 2 to 25 K, even with increasing magnetic field up to 15 T. Furthermore, when comparing the temperature dependence of its critical current (for  $H \parallel c$  at 0 T and 3 T) with other single crystals of iron-based superconductors or conventional superconductors in Figure 4(c)-(d), it is evident that  $\text{CaKFe}_4\text{As}_4$  displays a ultra-high self-field  $J_c$  around  $10^7$   $\text{A/cm}^2$  for  $H \parallel c$  and these characteristics remain robust up to 31 K in 3 T (up to a value of  $10^6$   $\text{A/cm}^2$ ). To further confirm these large  $J_c$  values, we have measured additional crystals, as shown in Fig.SM3 and Table SM1 that show very similar values to those presented earlier; a slight decrease in  $J_c$  is noticed as the thickness of the sample increases from  $2\mu\text{m}$  towards  $15\mu\text{m}$  (see Table SM1). These high  $J_c$  values of this stoichiometric clean superconductor with a relatively low residual resistivity ( $\rho_0 \sim 15.9\mu\Omega$  cm) and close to an optimum doping is surprising, as one would expect a high density of defects to achieve effective pinning and large critical currents.

In order to understand the nature of pinning, we study the temperature and field dependencies of vortex pinning force

density,  $F_P$ , given by  $F_P = \mu_0 H \times J_c$ , when  $H \perp J_c$ . According to the Dew-Hughes model<sup>21</sup>, if a dominant vortex pinning mechanism exists in certain temperature range, the normalized vortex pinning force density  $f_P = F_P / F_P^{max}$  at different temperatures should collapse into one curve, as shown in Fig.5. The resulting scaling law is given by  $f_P(h) = h^p(1-h)^q$ , where  $F_P^{max}$  is the maximum pinning force density; the indices  $p$  and  $q$  are two parameters whose values depend on the origin of pinning mechanism and  $h = H/H_{irr}$  is the reduced field, where the irreversibility field  $\mu_0 H_{irr}$  is estimated as the magnetic field at which  $J_c(H)$  reaches the background noise value, as defined in Fig. 3a.

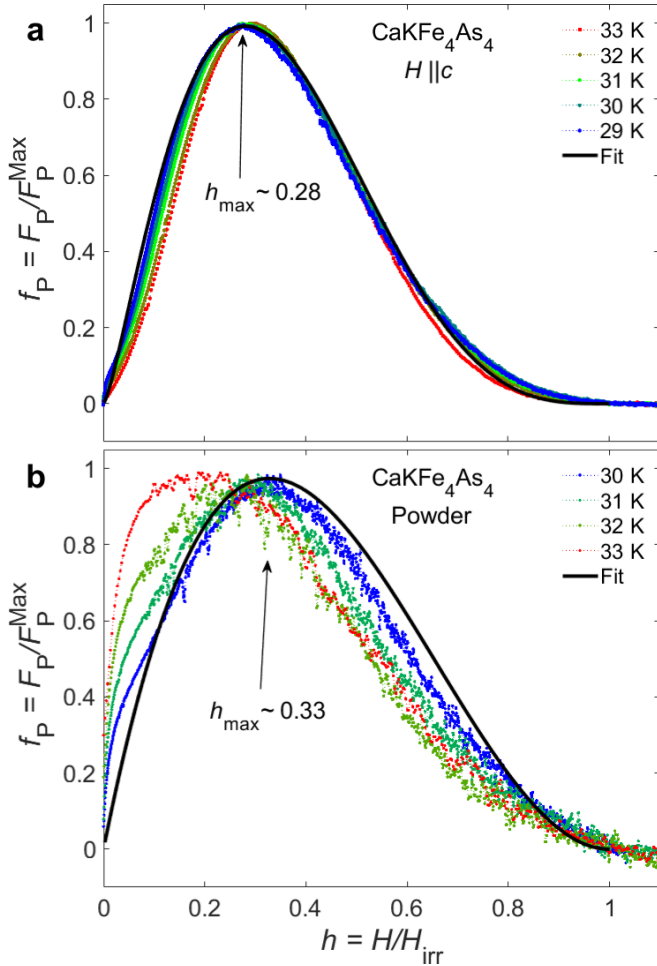


FIG. 5. The scaling density of the normalized pinning force density,  $f_P$ , for different temperatures as a function of the reduced field,  $h$ , based on the  $f_P(h)$  scaling function for (a) single crystal  $H||c$  and (b) powder sample of  $\text{CaKFe}_4\text{As}_4$ . The solid line represents the fitted curve and data measured at different temperatures scaled together with the maximum reduced magnetic field at  $h_{max} \sim 0.28$  for the single crystal in (a) and  $h_{max} \sim 0.33$  for the powder in (b).

Figure 5(a) and (b) shows the normalized pinning force density against the reduced magnetic field  $h$  of  $\text{CaKFe}_4\text{As}_4$  estimated at different temperatures for the single crystal ( $H||c$ ) and the powder sample. As  $H_{irr}$  exceeds the accessible magnetic field of 16 T at temperature below 27 K, only data closed

to  $T_c$  are presented ( $T \geq 29$  K). The calculated maximum pinning force density ( $F_P^{max}$ ) at 30 K reaches a value of up to  $9.58 \text{ GN/m}^3$  for the single crystal and a much reduced value of only  $0.0007 \text{ GN/m}^3$  for the powder sample. The normalized vortex pinning force density,  $f_P(h)$ , collapse onto on a single curve with a  $h_{max} \sim 0.28$  for the single crystal in Fig. 5a, indicating that a **dominant pinning mechanism exists** within this temperature range. For the powder sample (Fig.5(b)), the peaks in  $f_P(h)$  are much broader, due to its granular nature, however the  $h_{max}$  value is close to that of the single crystal. The fitting parameters to the  $f_P(h)$  give  $p = 1.4(1)$ ,  $q = 3.7(3)$  and  $h_{max} \sim 0.28$  for the single crystal  $H||c$  and  $p = 1.0(1)$ ,  $q = 2.1(1)$  and  $h_{max} \sim 0.33$  for the powder sample, respectively (Fig.5). In the Dew-Hughes model for pinning mechanism, a value of the  $h_{max} \sim 0.2$  corresponds to the surface pinning for normal center of core interaction, while  $h_{max} \sim 0.33$  corresponds to the point core pinning, also known as small size normal pinning<sup>21</sup>. Thus,  $h_{max} \sim 0.28 - 0.33$ , observed for  $\text{CaKFe}_4\text{As}_4$  suggests that vortex pinning can be caused by a mixture of the surface and point core pinning of the normal centres, similar to the  $\text{Fe}(\text{Se},\text{Te})$  systems<sup>49</sup>.

The obtained values of exponents  $p$  and  $q$  in  $\text{CaKFe}_4\text{As}_4$  listed in Table SM2 also support the idea that not a single model can describe the flux pinning mechanism<sup>21</sup> and the flux creep might have influence on the pinning force density. However, similar values of  $p$  and  $q$  have been reported for other unconventional superconductors, such as  $\text{FeTe}_{0.6}\text{Se}_{0.4}$  ( $h_{max} = 0.28$ ) and  $\text{YBa}_2\text{Cu}_3\text{O}_7$  ( $h_{max} = 0.33$ )<sup>49-51</sup> (see Table SM2). In the 122 iron-based superconductors, the pinning mechanism depends on the doping level and dopant and  $h_{max}$  tends to increase with increasing doping level<sup>16</sup>. The highest  $J_c$  is achieved when  $h_{max} \sim 0.40 - 0.45$  for the optimally-doped compounds<sup>16</sup>. Single crystal of  $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$  shows only small-size normal pinning ( $h_{max} = 0.33$ )<sup>21</sup>, whereas the values obtained for  $\text{BaFe}_{1.8}\text{Co}_{0.2}\text{As}_2$  single crystal<sup>16</sup> ( $h_{max} = 0.4$ ,  $p = 1.67$ ,  $q = 2$ ) are correlated to a dense vortex pinning nano-structure, likely due to the inhomogeneous distribution of cobalt ions. A high  $J_c$  is normally can be caused by a dense vortex pinning nano-structure mechanism. Violation of the scaling behaviour in the underdoped and overdoped 122 iron-based superconductors implies the existence of multiple pinning sources<sup>16</sup>. Stoichiometric  $\text{CaKFe}_4\text{As}_4$  shows superconducting behaviour equivalent to an optimally-doped superconductor<sup>16</sup>, but in the absence of inhomogeneities introduced by chemical substitution. Our scaling analysis implies that the proximity to a optimally-doping regime together with its mixed pinning mechanisms might be responsible for its high  $J_c$ .

Based on our magnetization studies, the vortex phase diagrams of  $\text{CaKFe}_4\text{As}_4$  for the single crystal (when  $H||c$ ) and the powder sample are shown in Figs. 6(a)-(b). There are five different characteristic fields separating different regions of the  $H - T$  vortex phase diagram.  $H_{min}$  and  $H_{sp}$  are the magnetic field located at the valley point and the second magnetization peak, respectively, in the  $J_c$  versus  $H$  plot, as defined in Fig.3(d). The irreversibility field  $H_{irr}$  is defined in Fig.3(d) and  $H_{c1}$  and  $H_{c2}$  have been discussed **previously** in detail in Fig. 2. In the powder sample, there is no second magnetization



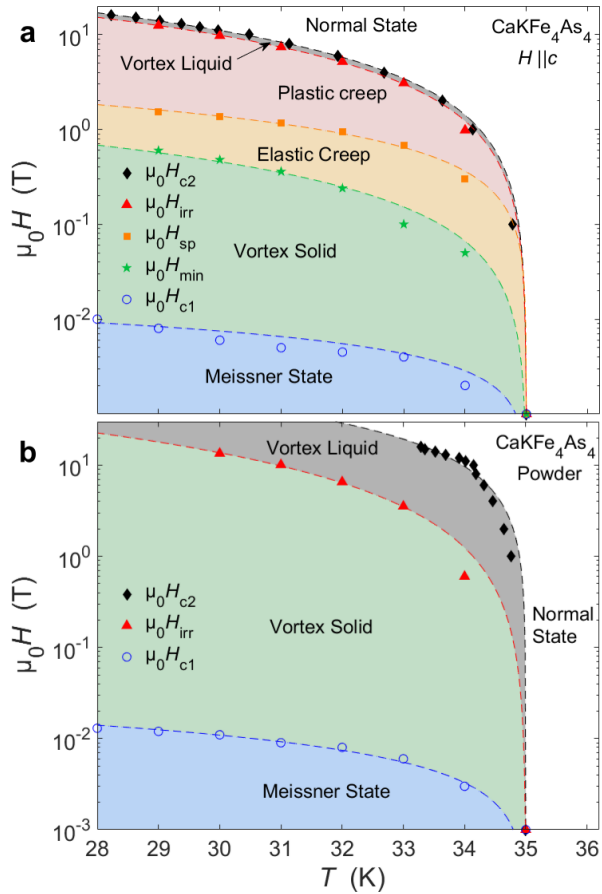


FIG. 6. The vortex phase diagram of a) a single crystal when  $H||c$  and b) polycrystalline sample of  $\text{CaKFe}_4\text{As}_4$ . The different magnetic fields separating different regions in the vortex phase diagram are defined in Fig.2 and Fig.3. The dash lines are fits using the empirical expression  $H(T) = H(0)(1 - T/T_c)^n$ .

peak. The irreversibility line,  $H_{irr}$ , is very close to  $H_{c2}$  in single crystals (Fig.6(a)) and this is a very important feature for  $\text{CaKFe}_4\text{As}_4$  to be technologically useful, as the irreversibility line demarcates the field at which vortex flow is unpinned and magnetic irreversibility sets in. On the other hand, the powder sample has  $H_{c2}$  a factor 4 larger than in single crystals and vortex liquid state is further extended, as shown in Fig.6(b). This suggests that grain boundaries increase the density of the pinning centers to give rise to larger upper critical fields, whereas the poor grain connectivity in powder is detrimental to supporting large critical currents, as they are significantly reduced.

Different regimes of vortex dynamics<sup>52,53</sup>, marked elastic creep and plastic creep, separated by the  $H_{sp}$  line, are shown in the phase diagram in Fig.6a. In general, the vortex phase diagram is determined by the competition between the elastic energy of the vortex lattice, the pinning energy and thermal energy<sup>16</sup>. The second magnetization peak effect is commonly associated with an order-disorder transition, which occurs at  $H_{min}$  when the pinning energy exceeds the elastic energy<sup>24</sup>. The motion of the vortices is governed by the elastic force in the elastic and plastic creep regions, and  $H_{sp}$  corresponds

to the threshold field of the elastic-plastic crossover. In the plastic creep region, as the magnetic field increases the vortex lattice softens and vortices are pinned more easily, resulting in an increase in  $J_c$ . As the magnetic field increases the pinning force and thermal energy affect the vortices significantly and the vortices can easily move from one pinning center to another in the vortex liquid state, which is above  $H_{irr}$ . In the case in which the pinning energy does not exceed the elastic energy in the entire field range, the SMP disappears, owing to the absence of an order-disorder transition, as in the case of powder sample in Fig. 6(b).

The vortex phase diagram of  $\text{CaKFe}_4\text{As}_4$  single crystal is very similar to that reported for the  $(\text{Ba,K})\text{Fe}_2\text{As}_2$  single crystal<sup>34</sup>, with  $H_{min}$  having almost the same value but  $H_{sp}$  and  $H_{irr}$  being higher in the 122 compounds. The temperature dependence of the different characteristic fields in Fig. 6(a) can be fitted using the empirical expression  $H(T) = H(0)(1 - T/T_c)^n$ , with  $n$  varying between 1.3(1) and 0.6(1). The obtained  $n$  value corresponding to the  $H_{min}$  curve is  $n=1.2(1)$ , similar to values reported for other iron-based superconductors<sup>36,54</sup>, whereas the  $H_{sp}$  and its corresponding exponent,  $n=0.83(6)$ , are much smaller than those found in other systems<sup>34,36,54</sup>. Furthermore, the vortex phase diagram of  $\text{CaKFe}_4\text{As}_4$ , delimited by the temperature dependence of  $H_{irr}$ , shows a high exponent value of  $n=1.3(1)$ , which cannot be understood within the framework of conventional superconductivity.

$\text{CaKFe}_4\text{As}_4$  is a stoichiometric rather clean superconductor exhibiting very high critical current values, rather unexpected, as a high density of defects is necessary to achieve such high values of  $J_c$ . However, recent STM studies find a disordered vortex lattice up to 8 T, with the vortices pinned at the locations where the superconducting order parameter is strongly suppressed due to pair breaking and the vortex core size decreases with increasing field which will give rise to very large upper critical fields<sup>55</sup>. The low-temperature vortex core size determined by STM is  $\sim 1.3$  nm, in reasonable agreement with the value estimated from upper critical field study of  $\sim 1.9$  nm, demonstrating the importance of point pinning in this material. Furthermore, the surface topography measured by STM exhibits steps on the surface due to structural defects, surface reconstruction and small stripes, which as possible sources of surface pinning. Other types of two-dimensional surface pinning may be influenced by the anisotropic layered structure and stacking faults that may occur between Fe layers along the c-axis.

The observed high  $J_c$  values are likely to be generated by intrinsic effects due to the ionic size variation that may occur in the crystallographic structure and the two-dimensional surface defects in  $\text{CaKFe}_4\text{As}_4$ . This structure has relevant differences compared to a 122 system, due to the absence of a glide plane and the positions of the Fe-As layers generating different Fe-As distances. The small structural defects can cause mean free path fluctuation induced pinning, which is an effective way to enhance  $J_c$ <sup>56</sup>. Furthermore, close to the optimal doping, as in the case of  $\text{CaKFe}_4\text{As}_4$ , the vortex core energy of the flux lines can be enhanced<sup>57</sup>. Thus, the high vortex core energy and the strength of the intrinsic depairing effect can be

key factors responsible for the extremely high  $J_c$  value in this compound. Generally, the intrinsically high depairing critical density is significantly reduced due to the presence of extrinsic effects, such as weak links and grain mismatch, as for our powder sample of  $\text{CaKFe}_4\text{As}_4$ .

#### IV. CONCLUSIONS

In summary, we have investigated the critical current densities and the vortex phase diagram of an optimally-doped stoichiometric superconductor,  $\text{CaKFe}_4\text{As}_4$ , both in single crystal and powder form. Interestingly, we find that this material exhibits extremely large  $J_c$  values up to  $10^8$  A/cm<sup>2</sup> at low temperatures, which are some of the highest values for a iron-based superconductor. The critical current density for both in-plane and out-plane orientations has a very weak temperature dependence in the temperature range up to 25 K. The magnetization curve shows very large hysteresis loops, **suggesting** strong flux pinning. We detect the fish-tail effect when  $H||c$ , similar to other optimally-doped 122 superconducting single crystals, but this effect is missing for the  $H||(\text{ab})$  orientation and the powder sample. The flux pinning force density suggests the existence of surface and the point core pinning of **the vortices** in low field regime. The extremely-high critical current density of  $\text{CaKFe}_4\text{As}_4$ , suggests that the vortex core energy of the flux lines can be enhanced in this optimally doped

superconductor and the reduced symmetry of As-Fe bonds could play an important role in pinning. Furthermore, in the powder sample the critical current densities are significantly reduced due to the weak grain connectivity, as often found in other iron-based superconductors. The ultra-high critical current densities and the vortex phase diagram of  $\text{CaKFe}_4\text{As}_4$  place this stoichiometric high- $T_c$  superconducting family as a realistic contender for practical applications.

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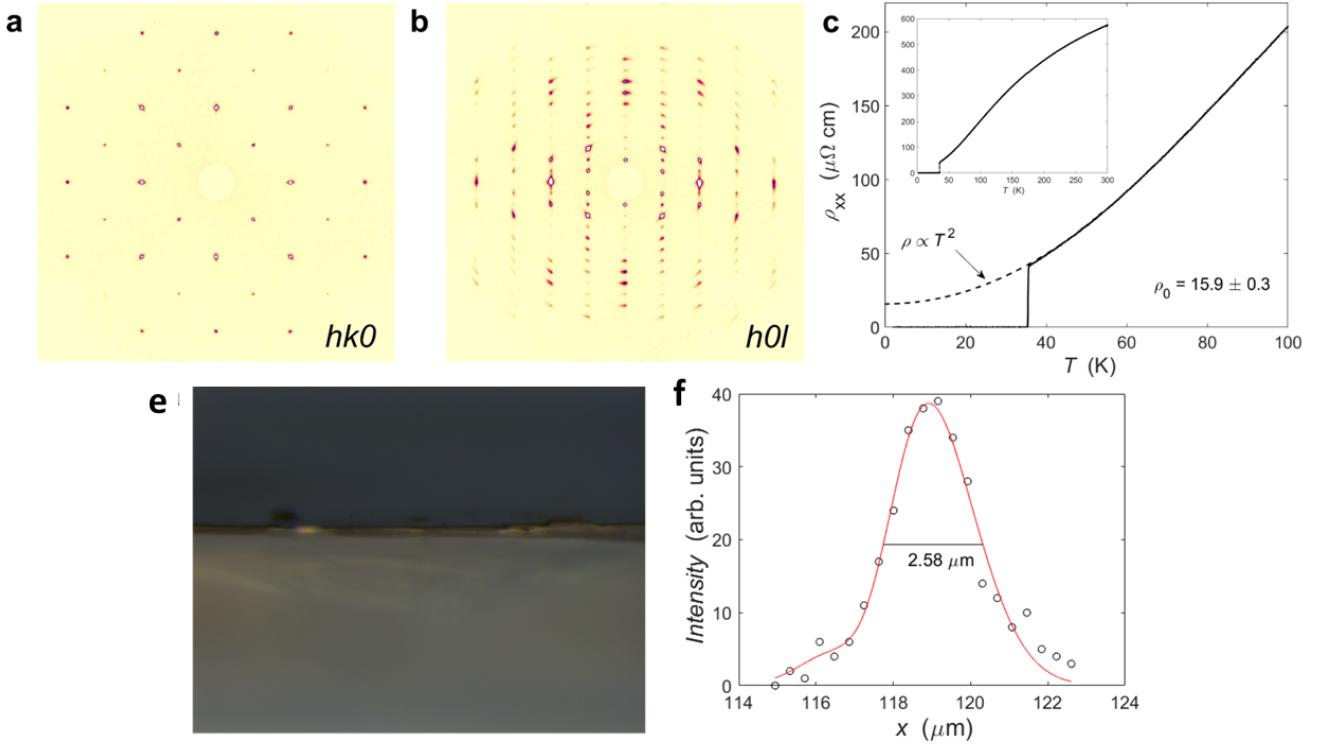


FIG. SM1. X-ray diffraction pattern of a) the  $(hk0)$  and b) the  $(h0l)$  plane for a single crystal from the same batch like the measured crystal. c) Resistivity data showing the very high quality of our single crystals giving an extrapolated low-temperature residual resistivity of  $\rho_0 \sim 15.9(3)\mu\Omega$  cm. The residual resistivity ratio, RRR (defined as  $\rho_{300\text{K}} / \rho_{36\text{K}}$ ) is  $\sim 14$ , in agreement with previous reports<sup>10</sup>, and together with the small superconducting transition width ( $\Delta T_c$ ) of 0.3 K, reflect the very high-quality of our single crystal. e) Optical image of the single crystal used in the magnetization studies. f) The estimation of the sample thickness from e) by fitting to a gaussian distribution of the intensity.

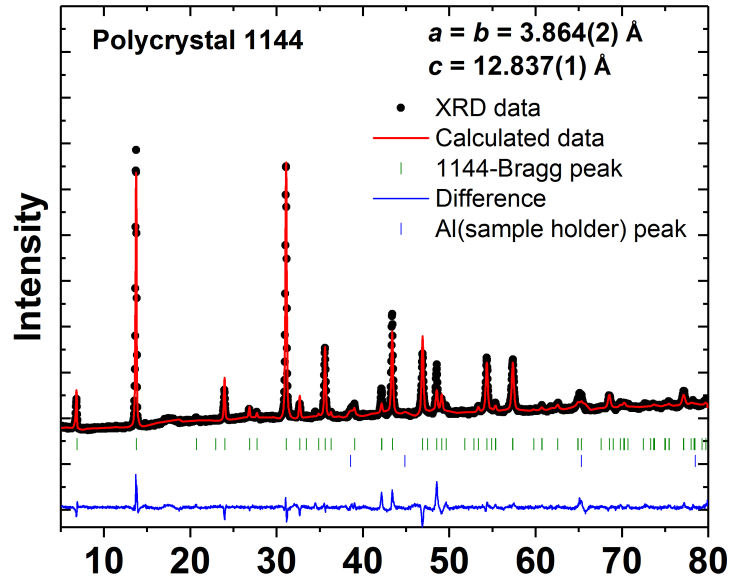


FIG. SM2. Powder x-ray diffraction patterns (XRD) and Rietveld refinement of the polycrystalline sample of  $\text{CaKFe}_4\text{As}_4$  used in this study. The X-ray data show that this is a single phase compound with the extracted lattice parameters closed to those reported for the single crystal ( $a = 3.8659$  Å,  $c = 12.884$  Å<sup>10</sup>).

TABLE SM1. The transition temperature,  $T_c$ , the slope of the upper critical field close to  $T_c$ , upper critical field,  $H_{c2}$ , and the lower critical field,  $H_{c1}^*$ , and the critical current density,  $J_c$ , extrapolated at zero field, of our single crystal (S1), an additional crystal S4 (thickness of 15  $\mu\text{m}$ ) and the powder sample of  $\text{CaKFe}_4\text{As}_4$ .

Parameters	Single Crystal	Single crystal	Polycrystalline
	$H  (\text{ab})$	$H  c$	
$T_c(\text{K})$	35.0(1)	35.1(2)	34.9(2)
$dH_{c2}/dT(\text{T/K})$	5.7	2.6	10.1
$H_{c2}(0)$	135	62	241
$H_{c1}^*(2\text{ K})(\text{T})$	0.1114	0.0220	0.0549
$J_c(\text{A/cm}^2)$ (5 K)(S1)	$1.9(2)\times 10^8$	$0.9(2)\times 10^7$	$4.4\times 10^3$
$J_c(\text{A/cm}^2)$ (5 K) (S2)	$0.8(2)\times 10^8$	$0.5(2)\times 10^7$	–
$J_c(\text{A/cm}^2)$ (5 K) (S4)	–	$0.4(2)\times 10^7$	–

TABLE SM2. Summary of the pinning force scaling parameters  $p$ ,  $q$  and  $h_{max}$ , for different superconductors.

Single crystal	$p$	$q$	$h_{max} = p/(p+q)$	Peak position
$\text{CaKFe}_4\text{As}_4$	1.4(1)	3.7(3)	0.28	0.28
$\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ <sup>34</sup>	1	2	0.33	0.33
$\text{Ba}_{0.68}\text{K}_{0.32}\text{Fe}_2\text{As}_2$ <sup>58</sup>				0.43
$\text{BaFe}_{1.8}\text{Co}_{0.2}\text{As}_2$ <sup>59</sup>	1.67	2	0.45	0.45
$\text{BaFe}_{1.85}\text{Co}_{0.15}\text{As}_2$ <sup>58</sup>				0.37
$\text{BaFe}_{1.29}\text{Ru}_{0.71}\text{As}_2$ <sup>60</sup>	1.95	2.5	0.44	0.45
$\text{BaFe}_{1.91}\text{Ni}_{0.09}\text{As}_2$ <sup>58</sup>				0.32
$\text{FeTe}_{0.7}\text{Se}_{0.3}$				0.27
$\text{FeTe}_{0.6}\text{Se}_{0.4}$ <sup>49</sup>	1.54	3.8	0.28	0.28
$\text{K}_x\text{Fe}_{2-y}\text{Se}_2$ <sup>61</sup>	0.86	1.83	0.32	0.33
$\text{YBa}_2\text{Cu}_3\text{O}_7$ <sup>50</sup>	2	4	0.33	0.33
$\text{NdBa}_2\text{Cu}_3\text{O}_{7-d}$ <sup>62</sup>	1.48	2.23	0.45	0.45
$\text{Sm}_{0.5}\text{Eu}_{0.5}\text{Ba}_2\text{Cu}_3\text{O}_7$ <sup>51</sup>	2.08	3.56	0.37	0.37

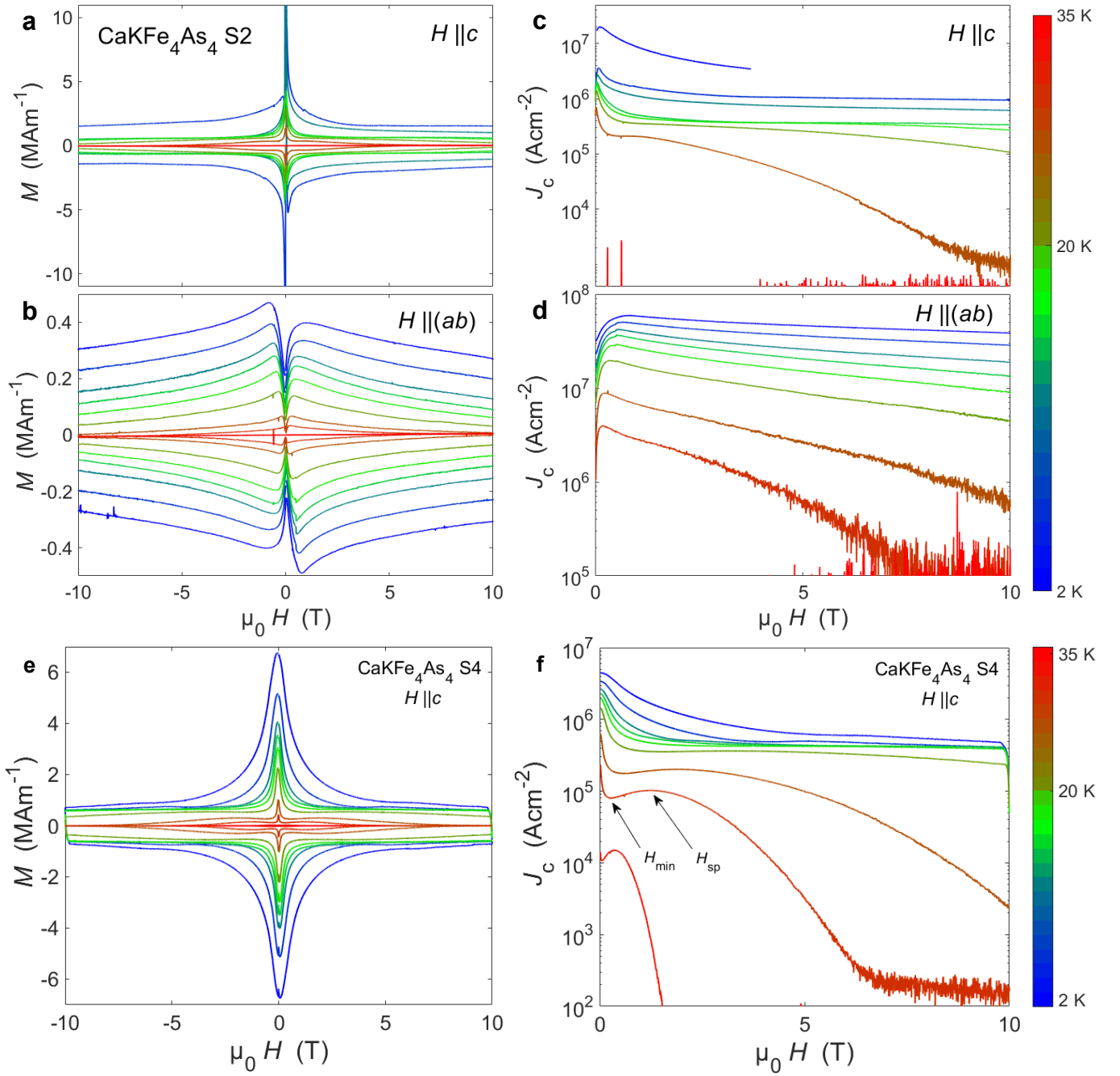


FIG. SM3. a), b) Magnetization and c), d) critical current density for a different single crystals, S2 (with thickness around  $12 \mu\text{m}$ ) measured with  $H \parallel c$  and  $H \parallel (ab)$ , respectively. e) Magnetization and f) critical current density for the single crystal S4 (with thickness around  $15 \mu\text{m}$ ) measured with  $H \parallel c$ .